

Application of a “Sealed Can Technique” and CR-39 detectors for measuring radon emanation from undamaged granitic ornamental building materials

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Abstract. Radon (^{222}Rn) is the most important cause of exposure to mankind due to natural radioactivity. Radon exhalation depends not only on the ^{226}Ra concentration in the material, but also on other factors such as mineralogy of the region, size and density of grains and porosity of the material. As building materials are one of the major sources of environmental radon, the radiological implications of the use of materials with great content of uranium must be always assessed. In this work, radon exhalation from several undamaged granitic building materials used as ornamental rocks or coating tiles is determined using the “sealed-can technique” and CR-39 solid state nuclear track detectors (SSNTD). Preliminary results for radon concentrations showed values of the same order that literature values, in a range from $99 \text{ Bq}\cdot\text{m}^{-3}$ to $1100 \text{ Bq}\cdot\text{m}^{-3}$. Further, the methodology will be validated with standard sources of ^{222}Rn .

1. INTRODUCTION

1.1. The Radon

The exposure due to inhalation of radon is the most significant factor of human irradiation by natural sources [1]. The three natural isotopes of radon, ^{222}Rn , ^{219}Rn and ^{220}Rn , each one from the radioactive natural series, respectively of ^{238}U , ^{235}U and ^{232}Th , [2], are all radioactive and alpha emitters.

Considering the short half-life of ^{220}Rn and ^{219}Rn and that ^{235}U represent only 0,71% of natural uranium [1], the greatest radiological concern focuses on the determination of the isotope of ^{222}Rn . The radon (radon, for simplicity, hereafter, means the isotope of ^{222}Rn) is a noble gas following the ^{226}Ra decay in the ^{238}U series. As a noble gas, radon does not interact with other elements and can easily emanate from the soil or rocks and concentrate indoors [3]. As uranium and radium are present in almost everything, like soil, rocks, water, building materials, etc., radon often is present in the surrounding air. However, the concentration levels are mainly determined by the following factors: floor covering (eg. pavement, buildings and vegetation), altitude, soil porosity and grain size, temperature, air pressure, moisture content of the soil, weather and seasons

1.2. Radon Measurements

Radon and its progeny can be detected by active and/or passive detection techniques [4]. In the present work, the radon and its daughters were detected by the passive technique of Solid State Nuclear Tracks Detectors (SSNTD) [4]. The operation of SSNTD detectors is based on the fact that heavy charged particles interact with the detector, leaving a latent track of the order of 50 to 100 Å, which, after specific chemical etching, can be visualized under an optical microscope [5].

To measure only radon, a diffusion chamber must be used, allowing solely the passage of radon, so the detector will register only the alpha emissions generated inside the diffusion chamber [4]. The radionuclides concentrations activities will be determined using a calibration factor, relating the track densities with activities.

1.3. Radon Exhalation

Exhalation is the number of radon atoms released from the surface of a specific material by emanation and diffusion.

Radon atoms are generated by alpha decay of ^{226}Ra with an initial energy of 86.0 keV and range in solid materials in the orders of tens of nm. Depending on the original site of the atom of ^{226}Ra in the structure and direction of recoil (Fig. 1), radon atoms can escape into the pores (micropores, cracks) (Fig. 1a), or be kept in mineral grains (Fig. 1b and Fig. 1c) [6]. For samples consisting of small grains, the pore spaces may not be enough to stop the recoil atom, consequently, some of the radon atoms can be implanted in neighborhood mineral grains, so, when released later, they are termed indirect recoil atoms [7]. When the pores are filled with water, the recoil atom is stopped and is free to diffusion; usually the moisture in the pores increases the emanation coefficient in relation to the pores filled with air. However, the atoms will diffuse more into the material if the pores are filled with air than the water [8]. This way, the exhalation of radon is quite complex and depends on many factors such as, for example, concentration and distribution of ^{226}Ra [8], grain size [7], moisture content [9] and mineralogical composition [6] of the material.

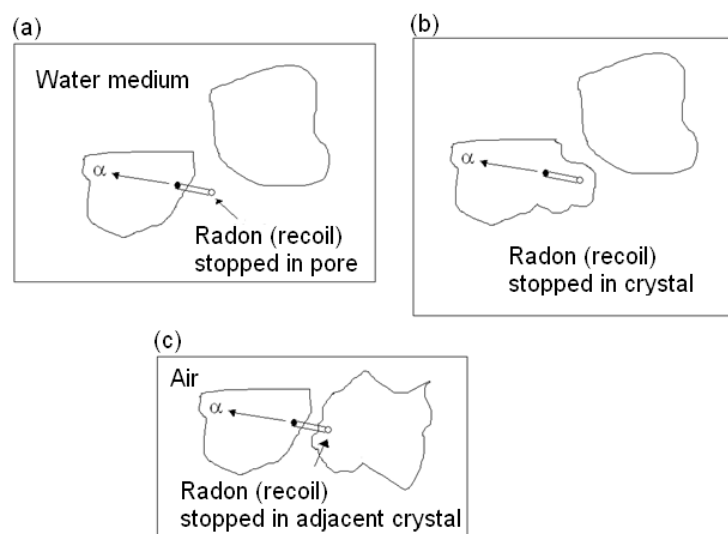


Fig. 1. Some processes of radon release

Usually, the indoors internal exposure index for radon is assessed through the activity concentration of ^{226}Ra . However, depending on the building material composition, it is possible that the real radon concentration in the air would be higher or lower when comparing with the ^{226}Ra content, so, it is of great importance the knowledge of radon exhalation.

2. MATERIAL AND METHOD

Six undamaged granite samples (Tab. 1) in the commercial form of 10 x 10 x 2 cm, were collected and directly exposed during three months in a sealed cylinder with CR-39 SSNTD.

Table 1 - Granite undamaged samples assessed in this work.

ID	Type	Trade name
GVI	Granite	Vermelho Itaipu
GIM	Granite	Imperador
GVV	Granite	Verde Vulcano
GSFG	Granite	Sea Foam Green
GBD	Granite	Bege Dunas
GPG	Granite	Paraná Green

2.1. Sealed Can Technique

The determination of radon and its progeny was performed by passive detection technique with solid state nuclear track detection (SSNTD), using the “sealed can technique”, [10-11-12]. Each sample was placed inside a plastic cylindrical container facing a CR-39 track detector into a diffusion chamber model NRPB/SSI [13] (Fig.2). The container was then sealed for approximately three months.

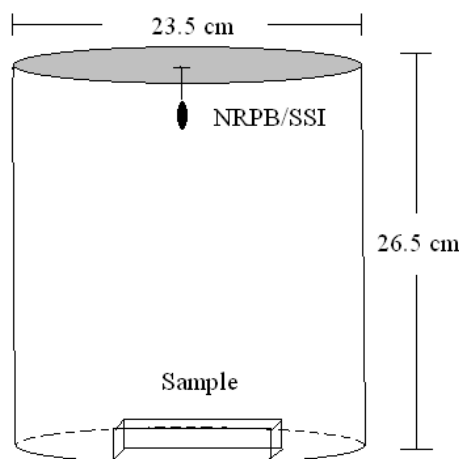


Fig. 2. Sealed Can Technique used in the present work

After exposure, the detectors were etched during 5.5 hours with a KOH 30% solution at 80°C [13], and readed under a Zeiss optical microscope [14].

The radon concentration C_{Rn} in ($Bq\ m^{-3}$) was determined by the following equation:

$$C_{Rn} = \frac{D}{F.t} \quad (1)$$

Where D is the tracks density in ($tracks.cm^{-2}$), t is the exposition time in (hours) and F is the calibration factor (detector efficiency) of $2.8 \pm 0.2\ tracks.m^3).(cm^{-2}.Bq^{-1}.h^{-1})$ [13].

The surface exhalation rate of radon E_x , in $Bq.m^{-2}.h^{-1}$, was calculated as follows:

$$E_x = \frac{CV}{A \left[T + \frac{1}{\lambda} (e^{\lambda T} - 1) \right]} \quad (3)$$

Where C is the integrated radon exposition measured by the CR-39 SSNTD ($Bq.m^{-3}.h$), V is the effective volume of the container, λ is the decay constant of radon (h^{-1}), T is the exposition time (h), and A is the area covered by the container (m^2) [12].

3. RESULTS

The radon activity concentration, the surface exhalation rate and the mass exhalation rate were calculated by the equations 1, 3 and 4 respectively and the results are show in the following figures,

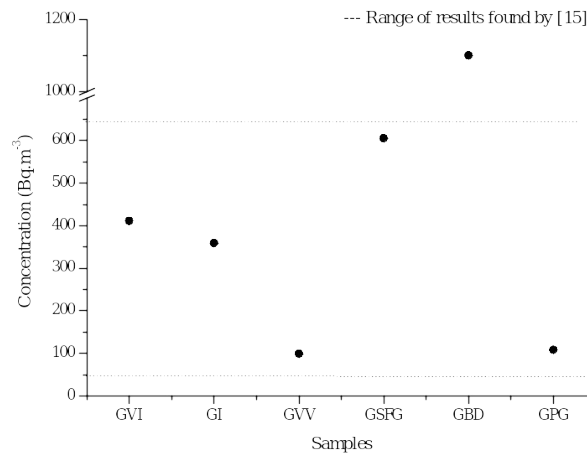


Fig. 3. Radon activity concentration ($Bq.m^{-3}$)

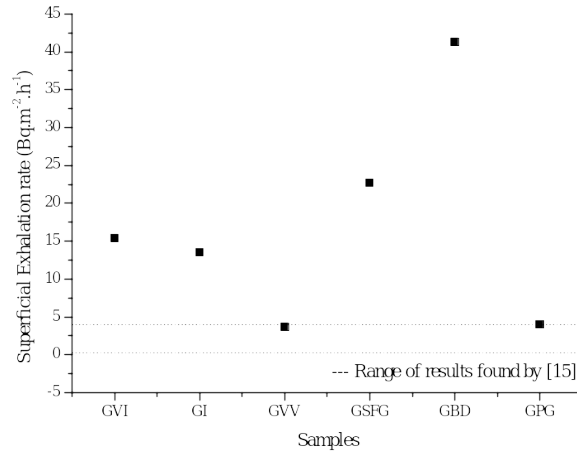


Fig. 4. Superficial Exhalation rate (Bq.m⁻².h⁻¹)

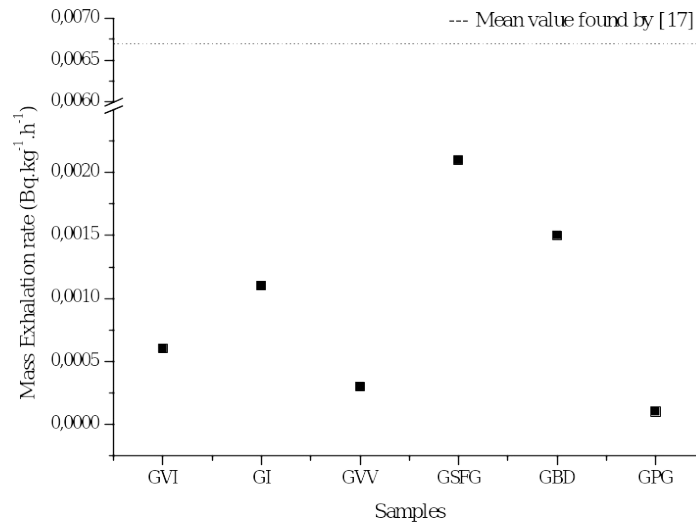


Fig. 5. Mass exhalation rate (Bq.kg⁻¹.h⁻¹)

The results of Fig. 3 show that the concentration of radon in all measured samples varies from 99 Bq.m⁻³ to 1100 Bq.m⁻³, in good agreement with the values for similar rocks found by Moura [15], ranging from 26 Bq.m⁻³ to 643 Bq.m⁻³ (dashed lines in Fig. 3). The surface exhalation rate (Fig.4) varies from 3.7 Bq.m⁻².h⁻¹ to 41.3 Bq.m⁻².h⁻¹. Similar values for the superficial exhalation rate were reported by Moura [15], ranging from 0.24 Bq.m⁻².h⁻¹ to 3.93 Bq.m⁻².h⁻¹ (dashed lines in Fig.4). The mass exhalation rate (Fig. 5) varies from 0.1 mBq.kg⁻¹.h⁻¹ to 2.1 mBq.kg⁻¹.h⁻¹. Kobeissi et alli [17] found a mean value of 6.7 mBq.kg⁻¹.h⁻¹ for mass exhalation rate for sands (dashed line in Fig.5), a higher value than the results found in the present work. However, considering that

radon exhalation is easier in sands than in granites, the granite results are to be lower.

4. CONCLUSIONS

The results for the radon concentration are in good agreement with literature for similar rocks. The measured surface exhalation rate was higher than the mass exhalation rate, allowing to conclude that the sample surface area has a major role in rising the exhalation rate, comparing with the sample mass. However, more measurements are needed in order to obtain suitable relationships between radon concentrations and surface areas for several materials.

Acknowledgements

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